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RADC-TR-89-90
Final Technical Report
August 1989

AD-A211 478

OPTIMIZATION, CHARACTERIZATION, AND IMPLEMENTATION OF A CW DYE LASER SYSTEM

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Griffiss Air Force Base, NY 13441-5700

89 8 17 111

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SECURITY CLASSIFICATION OF THIS PAGE

REPORT DOCUMENTATION PAGE

Form Approved OMB No. 0704-0188

1a. REPORT SECURITY CLASSIFICATION UNCLASSIFIED	1b. RESTRICTIVE MARKINGS N/A
2a. SECURITY CLASSIFICATION AUTHORITY N/A	3. DISTRIBUTION/AVAILABILITY OF REPORT Approved for public release; distribution unlimited.
2b. DECLASSIFICATION/DOWNGRADING SCHEDULE N/A	
4. PERFORMING ORGANIZATION REPORT NUMBER(S) N/A	5. MONITORING ORGANIZATION REPORT NUMBER(S) RADC-TR-89-90
6a. NAME OF PERFORMING ORGANIZATION Syracuse University 6b. OFFICE SYMBOL (If applicable) N/A	7a. NAME OF MONITORING ORGANIZATION Rome Air Development Center (OCTP)
6c. ADDRESS (City, State, and ZIP Code) Department of Chemistry Syracuse NY 13244	7b. ADDRESS (City, State, and ZIP Code) Griffiss AFB NY 13441-5270
8a. NAME OF FUNDING/SPONSORING ORGANIZATION AFOSR 8b. OFFICE SYMBOL (If applicable) NE	9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER F30602-81-C-0169
8c. ADDRESS (City, State, and ZIP Code) Bolling AFB Wash DC 20332	10. SOURCE OF FUNDING NUMBERS PROGRAM ELEMENT NO. 61102F PROJECT NO. 2305 TASK NO J9 WORK UNIT ACCESSION NO. P2

11. TITLE (Include Security Classification)

OPTIMIZATION, CHARACTERIZATION, AND IMPLEMENTATION OF A CW DYE LASER SYSTEM

12. PERSONAL AUTHOR(S)

Dr. Joseph Chaiken

13a. TYPE OF REPORT	13b. TIME COVERED	14. DATE OF REPORT (Year, Month, Day)	15. PAGE COUNT
Final	FROM May 87 TO Sep 87	August 1989	16

16. SUPPLEMENTARY NOTATION

N/A

17. COSATI CODES			18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number)
FIELD	GROUP	SUB-GROUP	
18	04		Intracavity, Dye Laser, Spectroscopy, NUCLEAR [illegible]
20	10		

19. ABSTRACT (Continue on reverse if necessary and identify by block number)

The overall project is to assess the applicability of intracavity laser absorption spectroscopy for in situ, non-instrusive monitoring of the desorption of barium containing species from operating thermionic cathodes. The goal was the implementation, characterization and optimization of a CW dye laser system with regard to its performance in an intracavity laser absorption apparatus. The purpose of the final report is to document the course of events associated with the task in the context of the overall mission of cathode evaluation and advanced approaches for predicting and improving cathode performance and lifetime. Four observations demonstrate the success of the project: 1) many unassigned absorption features due to the atmosphere (most likely due to molecular oxygen) were discovered in the vicinity of the barium absorption line. Plotting the active absorption strength of these features as a function of intracavity air pressure on a semilogarithmic scale yielded straight lines quantitatively in agreement with Beer's low type behavior. (see reverse)

20. DISTRIBUTION/AVAILABILITY OF ABSTRACT ☒ UNCLASSIFIED/UNLIMITED ☐ SAME AS RPT. ☐ DTIC USERS	21. ABSTRACT SECURITY CLASSIFICATION UNCLASSIFIED	
22a. NAME OF RESPONSIBLE INDIVIDUAL John F. McCalmont	22b. TELEPHONE (Include Area Code) (315) 330-4381	22c. OFFICE SYMBOL RADC (OCTP)

DD Form 1473, JUN 86 Previous editions are obsolete.

SECURITY CLASSIFICATION OF THIS PAGE

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Block 17 (Continued)

20 12

Block 19 (Continued)

2) The strength of any particular feature increased monotonically with generation time in agreement with the published accounts in the scientific literature.
3) Introduction of known absorbers into the ambient air gave measurable absorption features which could be assigned to known species. 4) Spectral output broadened as the generation time decreased. The observations are evidence that the ICLAS apparatus was performing in a manner comparable to the other accounts in the scientific literature.

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RADC FINAL REPORT
for Task #A-7-1905

Our goal was the implementation, characterization and optimization of a CW Dye Laser System(CWDLS) with regard to its performance in an Intracavity Laser Absorption Apparatus(ICLA). We were completely successful in meeting this goal. The purpose of this final report is to document the course of events associated with the task in the context of the overall mission of cathode evaluation and advanced approaches for predicting and improving cathode performance and lifetime. At this time two publications for the primary scientific literature are in preparation to follow this final report. Many technical details concerning the exact procedures and results will be presented in full via these other channels.

The overall project is to assess the applicability of intracavity laser absorption spectroscopy(ICLAS) for in situ, non-intrusive monitoring of the desorption of Barium containing species from operating thermionic cathodes. In the simplest case, the idea is to measure the absorption of light at 5535.5 Å which corresponds to a well-known and strong transition ($^1P^o$ <--- ^1S) of neutral Barium atoms. Any species which are desorbed and lead to the formation of Barium atoms in the vicinity of the dye laser modal volume will contribute to the absorption at this wavelength. It should be noted that in situ monitoring must necessarily account for atom-electron, molecule-electron, ion molecule, and ion-atom collisions, as well as other possibilities. The geometry is such that, as is shown in Figure 1, a cylinder is interrogated by the laser beam intracavity modal volume. It is anticipated that due to the overall low background pressure in the cathode test vehicle and the relatively low vapor pressure of Barium at room temperature, that the flow of possible absorbers being heated off the cathode surface is always in the effusive regime. Because of this, it should be possible to obtain absorption spectra at various positions as the laser beam is translated with respect to the cathode and thus obtain the spatial distribution of desorbed Barium containing species from the cathode surface. This distribution is expected to yield valuable insight into the geometric orientation of the sites on the cathode surface where Barium chemistry occurs. For example, it would be instructive to determine whether the various chemical processes occur mainly in pores or on flat and fully exposed surfaces. It is anticipated that the thermal desorption of Barium containing species which occurs during cathode activation will be indicative of the chemistry which results in an optimally emissive cathode surface. Correlation of the chemical and physical morphological changes which occur during activation with long term performance would constitute the basis for an advanced lifetime testing procedure.

While I have not been exhaustive in the above presentation of the underlying rationale for this project, it is clearly evident that demonstration of the ability to monitor the desorption of Barium containing chemical species is imperative, if a rational cathode lifetest procedure is going to be developed. There are probably a few different approaches to monitoring the desorption of Barium containing species which could be more or less successful, for example laser induced fluorescence(LIF) and FM spectroscopy(FMS), in addition to ICLAS. Without extensive prior knowledge of the true distribution of Barium containing species involved in the cathode chemical and morphological changes which occur during thermal activation, it is difficult to be definitive in a judgement of which method is most applicable for a cathode lifetest procedure. However, with some simple and plausible assumptions, ICLAS would seem to be the best choice. Consideration of the possibility of continuous internal calibration, the presence of copious thermionic emission and 1000-1200 K blackbody radiation, and the desirablilty of broad tunability of spectral coverage leads to a ranking, in order of decreasing usefulness, of ICLAS>LIF>FMS. It is quite possible that LIF could be a very competitive approach once some initial idea of the actual chemical species involved can be formed. FMS may well lead to a lower number density detection limit for most species but obtaining wide spectral coverage is less convenient and this also makes FMS less desirable for initial studies.

There has been considerable improvement in the theoretical description of the optical physics which we associate with the phenomena involved in ICLAS. The effort to develop a cathode test procedure using ICLAS initially was based on the particuliar procedure advanced by Brink. This procedure, which Brink has shown is equivalent to that of Harris and others, is also essentially equivalent to the method which we finally implemented. This equivalence would seem to be inadvertent but fortunate since the theoretical basis for the interpretation of ICLAS is now more complete and, it would seem, more realistic. This task was approached using the method theoretically described by Antonov and initially implemented in the laboratory by Antonov, Stoeckel, and Atkinson all independently of each other. Significantly, there are a few specific quantitative and qualitative tests which can be performed to demonstrate that the method is being implemented properly and these tests were in fact executed during the course of this task. Demonstration of the quantitative Beer's Law type relationship between a measured absorption and the number density of the absorber and similar but more qualitative observations regarding the relationship between the measured absorption and the generation time both clearly show that the ICLAS apparatus was performing as designed. By themselves, these simple observations constitute the basis of my claim that the task was executed successfully. What follows below is a brief description of the procedure which was adopted and the associated apparatus. Given this

description, a precise statement of the nature of the observations referred to above will be presented and will complete this final report.

An argon ion laser operating on all lines with an output of up to 4 W is directed through a beam reducing telescope before entering a high speed acousto-optic modulator(AO1). The modulated beam, which contains about 75% of the incident beam. encounters two turning mirrors and a focussing singlet before entering an ethylene glycol/Rhodamine 560 dye jet at Brewster's angle. This dye jet is placed in a standard three mirror folded optical cavity which also contains a high vacuum chamber. This chamber has Brewster Angle windows and can achieve a total pressure lower than 10^{-5} Torr *during* cathode activation. When dye laser action occurs, within 100 nsec of the turn-on of the Argon ion pump beam which itself requires 100 nsec, then roughly 7% of the confined coherent radiation is coupled out of the cavity. This radiation is spatially filtered and directed through a second acousto-optic modulator(AO2). The modulated portion of this beam is directed through a light collection system before being spectrally dispersed by a .8 m spectrograph with a 1200 grooves/mm grating. The light was detected by an optical multichannel analyzer(EGG OMA-II) with 25 μm wide diodes corresponding to each pixel. This suggests a calibration of roughly .065 Å /pixel. This system was typically operated in second order with the position of a reference Ba emission line from a hollow cathode lamp being reproducible to within ± 4 pixels. Under our light collection conditions, and based on the number of grooves in our grating, the best theoretical resolution we could attain would be about 50000. In practice, the system probably only achieved around 35-45000. Substantial improvement is attainable by minimization of mechanical instabilities in the light collection system. During the course of this task, the dye laser operation and radiation was repeatedly characterized with respect to spectral bandwidth, temporal stability, polarization properties, spatial mode structure and other properties which we could observe.

The operation of the acousto-optic modulators and the overall procedure can be explained with reference to the timing diagram depicted in Figure 2. A typical measurement sequence begins with the turning on of the argon ion pump beam. Depending on the intensity of the pump beam, the quality of the optical cavity, the condition of the dye/solvent jet, ie. the various factors which influence the threshold pumping required to initiate and sustain lasing action, the dye laser begins to emit detectable radiation through its output coupler at some time after pumping begins. At this time, termed the "generation time" and arbitrarily measured from the time that the pump beam reaches its 90% of final level, a 10 μsec "slice" of dye laser output is directed into the light collection/analysis system by the AO2. This overall approach to sampling the

dye laser output, termed the "quasi-CW method", has a very simple interpretation and leads to Beer's Law type expressions relating a measured absorption to the generation time, the number density of the absorber and a standard absorption coefficient. This absorption coefficient is well-known for the particuliar Barium transition being monitored. The generation time and the cavity length can be combined with the generation time to express a generalized path length for inclusion into the Beer's Law type expression. Lengthy statistical arguments have been employed by Antonov to establish the hypothesis that light coupled out of the dye laser cavity during the generation time reflects the presence of absorbers and refracters in the laser cavity with good signal to noise whereas after the dye laser output has reached "steady state" , the spectral distribution of the emitted radiation is more sensitive to quantum statistical fluctuations in the intracavity radiation field causing the effective attainable signal to noise to be reduced with respect to the effects of intracavity absorbers and refracters.

Among the observations which demonstrate the success of our labors note the following. 1) Our earliest attempts to detect Barium absorption were hampered by the presence of many absorption features in the vicinity of the one we desired to monitor. Occurring in a spectral region previously believed to be transparent for the constituents of air, we conclusively showed that all these interfering absorptions were in fact due to air and most likely molecular oxygen. Modification of the vacuum chamber to exclude more air from the intracavity modal volume caused an adequate reduction in the strength of the interfering absorptions. Plotting the actual absorption strength for a number of these still unassigned features as a function of intracavity air pressure on a semilogarithmic scale yielded straight lines quantitatively in agreement with a Beer's Law type behavior. 2) The strength of any particular feature increased monotonically with generation time in agreement with the theoretical predictions of Antonov and the experimental verifications of Stoeckel and others. 3) Introduction of known absorbers into the ambient air near the edges of the dye laser cavity gave measurable absorption features which could plausibly be assigned to the known species. 4) The dye laser produced a substantially broader spectral output as the generation time was decreased. 5) The tunability of the dye laser was a strong function of the presence of absorbers and refracters in the cavity. The laser could not operate centered at wavelengths which corresponded to absorption features without the use of an intracavity etalon. <u>These observations all support the statement that the ICLAS apparatus was performing in a manner comparable to the other accounts in the scientific literature</u>. The dye laser was therefore well characterized, optimized and fully implemented in the ICLAS apparatus.

Although there were many aspects of this research in which I contributed, I will list a few items which give some idea of my role in this project. It was my decision to employ the newer ICLAS methodology of Antonov and Stoeckel. The choices involving instrumentation for implementation of this different procedure were mine. I was mostly concerned with the underlying understanding of the spectroscopic aspects of this project, the data manipulation of the raw measurements to transform it into absorption data, and the interpretation of the observations and elimination of experimental artifacts. These were large aspects of the overall project and involved many subtasks. Three Syracuse University graduate students contributed to this project. Jeanne Hossenlopp, Dan Rooney and Brian Samoriski wrote computer programs for simulating the dye laser cavity performance and searching the extensive lists of known Barium and molecular oxygen spectroscopic states, searched the literature for ICLAS relevant reports and cathode relevant publications, and designed and supervised the construction of the electronic and mechanical additions which were made during the course of the task. Having stated my and my Syracuse University research group's main contributions, it is necessary to also state that this project could not have been accomplished without the competent, enthusiastic and determined assistance of RADC personnel; Lt. John McCalmont, Dr. Ed Daniszewski, Capt. Andy Chrostowski and Mr. Bohdan Kwasowsky. The majority of the actual hands on experimentation was executed by these able colleagues and, in an experiment which is as sensitive and delicate as ICLAS, this is a formidable accomplishment. I often functioned as a telephone consultant with these talented coworkers and this is a situation which simply would not have been possible if my RADC counterparts were not able to quickly and clearly learn the associated practical and theoretical background associated with the primary goal of implementing and optimizing the dye laser.

Regarding the overall goal of this project, i.e. the development of an advanced cathode lifetest/prediction procedure, it would seem that ICLAS can be employed with a very good probability of success. Many attempts were made to detect neutral Barium atoms desorbed from a cathode and other cathode-like materials and the success which is evident in our results strongly suggest that ICLAS possesses more than adequate sensitivity and versatility to serve as a basic evaluation technique for an accelerated cathode lifetest procedure. I note for the record that much spectroscopic work remains to be done, and that the physical chemistry/kinetics of the cathode activation procedure is clearly very complex, but there is ample reason to be optimistic that continued work in characterizing the reaction products which are desorbed from actual cathodes during activation and operation using ICLAS will lead to a viable cathode lifetest procedure.

Figure 1. **INTRACAVITY LASER ABSORPTION APPARATUS**

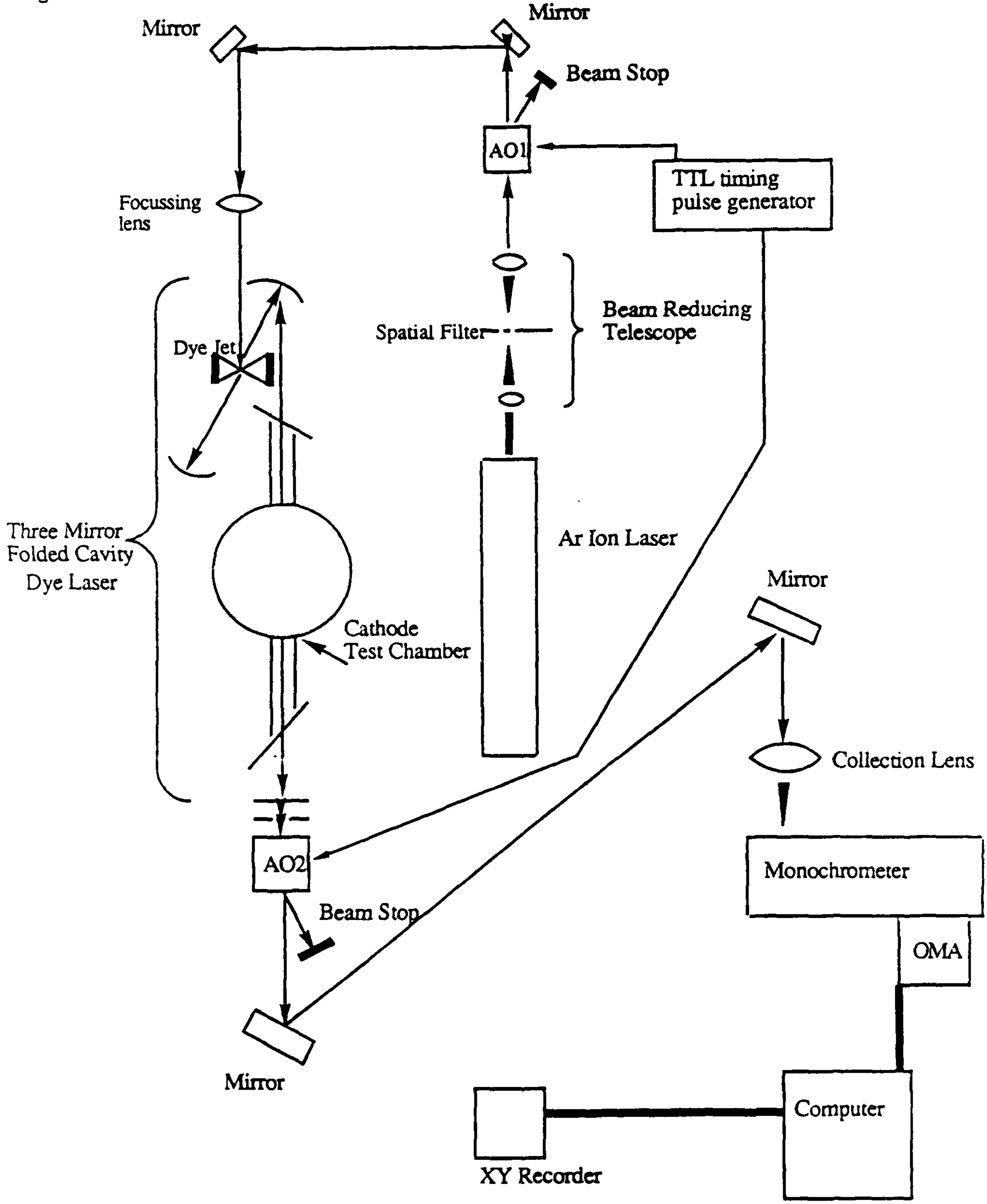
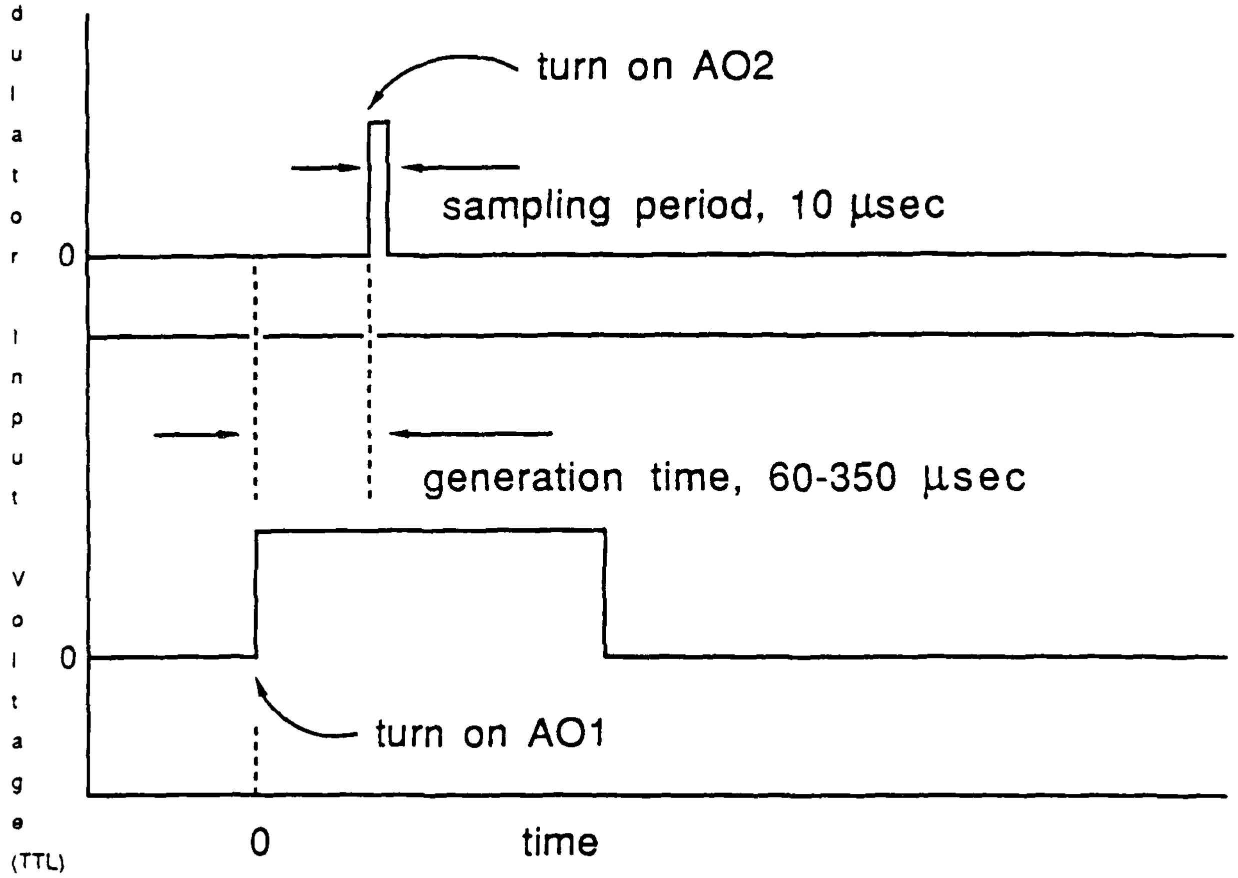


Figure 2 Quasi-CW Method Timing Diagram



Timing Diagram Showing Operation of the Acousto-Optic Modulators

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